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MiReCOL: REMEDIATION OF SHALLOW LEAKAGE FROM A CO_2 STORAGE SITE

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Abstract

Within the EU-funded project MiReCOL (Remediation and Mitigation of CO_2 Leakage; project number 608608), a comprehensive review was undertaken of techniques available for the remediation of leakage of CO_2 to the near surface environment, here defined as the depth range of typical remediation techniques used by the pollution clean-up industry. The review drew from existing relevant fields of experience such as the remediation of groundwater pollution; the remediation of industrial waste; CO_2-EOR, natural gas storage sites; the geothermal energy industry; the construction of dams (as barriers to subsurface fluid flow); pilot scale CCS projects and natural analogues.

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Keywords: remediation; CO_2 leakage; shallow subsuface; Mirecol; cost

1. Introduction

This paper provides a summary of near-surface CO_2 leakage remediation methods, including an assessment of their effectiveness. The near surface environment is defined as the depth range of typical remediation techniques used by the pollution clean-up industry, rather than by the hydrocarbon industry. The techniques for the remediation of a CO_2

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leak are taken from relevant fields that provide analogues for the CO_2 storage industry and facilitate the evaluation of mitigation and remediation procedures.

The applicability of each CO_2 leakage remediation method in the near surface environment, the ease of implementation and the associated costs for each method were compiled to produce a summary table to indicate the probable role different remediation techniques could play in the near-surface environment. The results indicate that a wide range of remediation techniques may be used for near surface CO_2 remediation and that any remediation strategy will need to be site specific to be effective.

1.1 Previous reviews of remediation technologies and methodologies

There is a recent review of the remediation of the leakage of CO_2 from a CO_2 storage site [1] which is broad in scope, but includes the remediation of near surface leakage as part of a wider review. Other relevant reviews include:

- vadose zone remediation [2];
- early detection of CO_2 leakage and remediation [3];
- the IPCC report from 2005 [4];
- a model of CO_2 leakage specifically designed for the near-surface [5];
- the very comprehensive review of the IEA GHG [6];
- a very useful review of natural CO_2 emissions sites, as a part of the UK QICS project [7];
- state of the art monitoring methods to evaluate CO_2 storage site performance [8].

Outside the fledgling CCS literature, there is little published concerning the remediation of CO_2 leakage. The journal 'Remediation' which, as the title suggests, is dedicated to environmental clean-up technologies, techniques and costs, appears to have no papers specifically concerning the remediation of leaks of CO_2. No text book appears to consider the problem. A more complete version of this review can be found on the MiReCOL website (http://www.mirecol-co2.eu/), including a comprehensive reference list that cannot be included here.

2. Methods

A literature review was undertaken. Techniques considered suitable for CO_2 leakage remediation were studied from other relevant fields, as there is relatively little experience of remediation of shallow CO_2 leaks. The relevant fields were:

- The control of groundwater pollution, especially potable water – in near surface environments. CO_2 in the gas phase has a similar density to some volatile organic compound (VOC) vapours, which are a common pollutant that is considered in remediation. However, it should be noted that CO_2 is non- toxic at low concentrations and is generally sourced from below the rock / soil matrix that requires remediation [9];
- Oil / gas operations (including EOR / CO_2-EOR) including both routine and acute incident scenarios – there are no recorded instances of leakage to the surface that did not involve boreholes;
- Natural gas storage projects [3];
- CO_2 production for EOR (e.g. the blow-out at Sheep mountain, Colorado, USA [6]);
- Natural analogues for surface leakage (e.g. Crystal Geyser, Utah, USA);
- Geothermal power in high-CO_2 regions (e.g. Torre Alfina, Italy);
- The grouting of the foundations of dams (for water storage);
- Pilot-scale and proposed industrial-scale carbon capture and storage (CCS).

3. Results

Given that the focus of this paper is the near surface environment, then there are a number of factors which make this environment different from that being considered for the deeper subsurface, which is the realm of the hydrocarbon industry:

- Low to very low water salinity (typically << 35 ppt NaCl, i.e. seawater equivalent);
- Higher water flow rates;
- CO_2 in gas phase, possibly present as hydrates;
- Natural fractures may be open due to low confining pressure;
- In an active sedimentary basin:
    - unconsolidated, uncemented sediments;
    - very high porosity and permeability (> 20 % and Darcy scale permeability);
    - low capillary entry pressure;
    - biological activity including:
        - biodegradation of hydrocarbons;
        - formation of kerogen and biogenic methane;
    - lack of structures (traps) to collect leaked CO_2;
    - lack of (active) faults as pathways for leakage;
    - presence of polygonal clay shrinkage cracks.

The possible pathways for the leakage of CO_2 in the near surface are similar to those associated with leakage at depths that are typical of those encountered by the hydrocarbon production industry:

- Boreholes – both abandoned and active;
- Faults and fractures, including both those sufficiently large for resolution using seismic imaging, and those too small for seismic resolution;
- Matrix rock porosity within lithologies such as sandstones and limestones.

3.1 The aims and objectives of remediation

The aims and objectives of remediation of leaked CO_2 will vary from site to site, according to the likely impacts and consequences. Generally, the aims will include:

- To stop the source of the leakage – in the context of the near surface, the leak is almost certainly sourced from a much a deeper storage reservoir, and mitigation at depth is probably more appropriate;
- To reduce the mobile free phase CO_2, to limit the continued growth of the leakage plume, i.e. to prevent the spread of the contamination [10];
- To delay the spread of a plume or dissolved CO_2, either while plans are drawn up for permanent remediation, or while legal action takes place to determine who is going to pay for remediation;
- To remove CO_2 from the aquifer in both gas and aqueous phase, both to recover the CO_2 for disposal and to restore the aquifer back to pre-contamination conditions [10];
- To minimise the decrease in pH from the formation of carbonic acid. Minimising the drop in pH may indirectly decrease the amount of secondary contamination from the CO_2 leakage caused by the mobilisation of heavy metal ions (e.g. [10,11]);
- To directly reduce the concentration of mobilised toxic metals to either background levels, or to levels acceptable to relevant legislation.
- To reduce the concentration of hydrocarbons that may be mixed with, or dissolved in, the leaking CO_2, especially if the primary storage reservoir is a depleted gas or field, or a depleted oil field with a high proportion of light oil that can volatilise into the free CO_2 phase;
- Prevent the CO_2 from reaching the surface, to avoid payment of fines or the return of credits for the avoidance of CO_2 emissions;

- Prevent the CO_2 from reaching habitations or other sensitive locations ('receptor' in pollution control terminology).

3.2 Classification of remediation techniques

There are a number of different remediation technologies suitable for the near surface remediation of CO_2 leakage, which can be classified by:

- Objective of the technology (containment or treatment);
- Process involved in the remediation (physical, chemical, biological or thermal);
- Location of the remediation process (in situ or ex-situ).

3.2.1 Containment versus treatment

Containment prevents the spread of the CO_2 without necessarily removing or degrading the contamination. Treatment transforms the CO_2 into less toxic, or non-toxic concentrations. Containment is typically cheaper, can be used until a more efficient clean up technology becomes available, can provide a means of evaluating the potential for natural attenuation processes to degrade the CO_2 and can present a lower overall risk as CO_2 exposure can be minimized [12]. Many remediation technologies will involve both containment and treatment.

3.2.2 In-situ or ex-situ remediation

Here it is important to highlight the distinction between the application of the remediation technology versus the location of the remediation treatment, for example in pump and treat the pumping is in-situ but the treatment of the CO_2 contamination is ex-situ [13].

3.2.3 Active or passive technologies

Passive containment refers to treatment systems that clean up the CO_2 contamination without the need for energy input for the treatment process to be effective. In contrast, active technologies require further enhancements or energy inputs to achieve the required level of clean up [14]. Active systems are generally more expensive than passive systems.

These are a number of remediation techniques available for the clean-up of CO_2 at shallow burial depths (Table 1).

Table 1. Summary of the shallow surface CO_2 remediation technologies available.

Remediation	**Remediation Technique**	**Containment or treatment**	**in-situ or ex-situ**	**Active or passive**
Fluid control measures	Pump and treat	Treatment	In-situ technology, ex-situ treatment	Active
	Pump and treat with cap	Containment and treatment	In-situ technology, ex-situ treatment	Active
	Water injection	Treatment	In-situ technology, ex-situ treatment	Active
	Hydrodynamic isolation	Treatment	In-situ	Active
	Air stripping	Treatment		Active
	Hydraulic barrier	Containment and	In-situ	Active

		treatment		
Cut off wall (unconfined aquifer)	Cut-off wall / slurry wall	Containment	In-situ	Passive
	Two-phase diaphragm wall	Containment	In-situ	Passive
	Composite diaphragm wall	Containment	In-situ	Passive
	Interlocking bored-pile diaphragm wall	Containment	In-situ	Passive
	Installation of thin wall and sheet pile into the soil	Containment	In-situ	Passive
	Injection permeation grouting	Containment	In-situ	Passive
	Jet grouting	Containment	In-situ	Passive
	Frozen wall	Containment	In-situ	Passive
	Bio barrier	Containment	In-situ	Passive
	Water control agent	Containment	In-situ	Passive
	High strength rigid set material	Containment	In-situ	Passive
	Organic polymer sealant	Containment	In-situ	Passive
	Super absorbent crystals	Containment	In-situ	Passive
	Granular activated carbon	Treatment	In-situ technology, ex-situ treatment	Active
Cut off wall - Fractured aquifer	Grout curtain	Containment	In-situ	Passive
Permeable reactive barriers (treatment walls)	Sorption barriers	Treatment	In-situ	Passive
	Ionic species removal	Treatment	In-situ	Passive
	Microbes	Treatment	In-situ	Passive
	Carbonation stabilisation	Treatment	In-situ	Passive
	De-acidisation	Treatment	In-situ	Passive
Soil Zone remediation	Soil vapour extraction	Treatment	In-situ technology, ex-situ treatment	Active
	Air sparging	Treatment	In-situ technology, ex-situ treatment	Active
	Bioslurping	Treatment	In-situ technology, ex-situ treatment	Active
	De-acidise soil	Treatment	In-situ	Passive
	Thermal treatment	In-situ technology, ex-situ treatment	In-situ technology, ex-situ treatment	Active
	Capping	Containment	In-situ	Passive
	Gas collection trench	Treatment	In-situ	Passive
	Ecosystem restoration	Treatment	In-situ	Active
Bioremediation	Bioremediation of low pH	Treatment	In-situ	Passive

	groundwaters			
	Bioremediation of CO_2	Treatment	In-situ	Passive
	Bioremediation of toxic metals	Treatment	In-situ	Passive
	Bioremediation of hydrocarbons	Treatment	In-situ	Passive
	Natural attenuation	Containment	In-situ	Passive
Buildings	Passive vapour intrusion mitigation	Treatment	In-situ	Passive
	Passive / active sub slab venting	Treatment	In-situ	Passive
	Active vapour intrusion mitigation – subsurface pressurisation	Treatment	In-situ	Active
	Block wall depressurisation	Treatment	In-situ	Passive
	Active ventilation	Treatment	In-situ	Active
	Passive ventilation	Treatment	In-situ	Passive
	Demolish and rebuild to suitable standards.	Treatment	In-situ	Active

4. Discussion

The applicability of the potential remediation techniques listed in the previous section was assessed, using literature data. The probable role was assessed in terms of:

- Practicality of application to CO_2 contamination. Is there an established CO_2 remediation application (or at least a reasonable expectation that the application would successfully remediate CO_2) or is it a potential but untested possibility;
- Ease of implementation of the remediation technology – is it an easy deployed in-situ technology with passive maintenance or a technology that requires significant ground works and implementation infrastructure and active maintenance;
- Cost – reasonable or so expensive it prohibits the use of the technology.

The techniques were hence classified against the categories listed in Table 2.

Table 2. Summary of the probable role grading

Probable role	CO_2 applicability	Easy of technology implementation	costs
Likely	Proven / established CO_2 applicability	Relatively straightforward technology application	Reasonable
High	Potentially applicable to	Relatively straightforward technology	Reasonable

intermediate	CO_2 contamination	application	
Intermediate	Potentially applicable to CO_2 contamination	Complex technology application	High
Minor	Potentially applicable to CO_2 contamination	Complex technology application	Very high
Unlikely	not directly applicable to CO_2 contaminations	Complex technology application	Very high

Note that, as costs are generally specified in the literature, and in any case are specific to the leakage scenario being remediated, that cots are classified only as 'reasonable' to 'very high'. More specific costing was not practicable.

Table 3. Summary assessment of the probable role each of the remediation techniques with regards to CO_2 remediation.

Remediation	Remediation Technique	Probable role	Improvements / comments
Fluid control measures	Pump and treat	**Likely**	Larger plumes may require horizontal wells and longer remediation times.
	Pump and treat with cap	**Likely**	Cost will depend on extent of cap
	Water injection	**High Intermediate**	Useful short term to reduce concentration of CO_2, but residually trapped CO_2 remains.
	Hydrodynamic isolation	**Likely**	Stabilises CO_2 plume
	Air stripping	**Likely**	Process is quick and relatively cheap
	Hydraulic barrier	**High Intermediate**	Works if aquifer is not very permeable and location of leak is known
Cut off wall (unconfined aquifer)	Cut-off wall / slurry wall	**Intermediate**	High costs depending on length of wall, risk of wall leakage and degradation. Only provide partial containment and further clean up technologies needed
	Two-phase diaphragm wall	**Intermediate**	High costs depending on length of wall, risk of wall leakage and degradation. Only provide partial containment and further clean up technologies needed
	Composite diaphragm wall	**Intermediate**	High costs depending on length of wall, risk of wall leakage and degradation. Only provide partial containment and further clean up technologies needed
	Interlocking bored-pile diaphragm wall	**Intermediate**	High costs depending on length of wall, risk of wall leakage and degradation. Only provide partial containment and further clean up technologies needed
	Installation of thin wall and sheet pile into the soil	**Intermediate**	High costs depending on length of wall and risk of sheet material corrosion
	Injection permeation grouting	**Intermediate**	Leakage risk through permeability gaps. Only provide partial containment and further clean up technologies needed

	Jet grouting	**Intermediate**	Leakage risk through permeability gaps. Only provide partial containment and further clean up technologies needed
	Frozen wall	**Unlikely**	Requires the active (powered) circulation of refrigerant coolant or liquid nitrogen
	Bio barrier	**Intermediate**	Technology untested in situ for CO_2, costs and application low.
	Water control agent	**High intermediate**	Technology available and low cost. Resistance to CO_2 untested.
	High strength rigid set material	**High intermediate**	Technology available and low cost. Resistance to CO_2 untested.
	Organic polymer sealant	**High intermediate**	Technology available and low cost. Resistance to CO_2 untested.
	Super absorbent crystals	**High intermediate**	Technology available and low cost. Resistance to CO_2 untested.
	Granular activated carbon	**Likely**	Process is relatively quick and cheap but will depend on CO_2 concentration or volume
Cut off wall - Fractured aquifer	Grout curtain	**Likely**	Boreholes ideally orientated to intersect as many fractures as possible, fracture permeability important and can be enhanced through hydraulic fracturing. Grouting materials need to be CO_2 resistant
Permeable reactive barriers (treatment walls)	Sorption barriers	**Likely**	Sorption materials need to be CO_2 specific. Over time reactive materials become less effective at removing CO_2 and the contaminated reactive material needs to be removed and replaced with fresh material.
	Ionic species removal	**High Intermediate**	Established procedure to clean up the trace elements potentially mobilised by the CO_2 contamination
	Microbes	**Intermediate / minor**	A cheap option but CO_2 specific microbes that will be in optimum conditions are hard to establish
	Carbonation stabilisation	**Intermediate / minor**	A cheap option but carbonation rates are hard to establish
	De-acidisation	**Likely**	Established cheap technology
Soil Zone remediation	Soil vapour extraction	**Likely**	Potential to be used in conjunction with containment treatments.
	Air sparging	**High Intermediate**	CO_2 will follow high permeability pathways so initial recovery rates high but will fall off as recovery is limited to diffusion. Potential to be used in

			conjunction with containment treatments
	Bioslurping	**High Intermediate**	CO_2 will follow high permeability pathways so initial recovery rates high but will fall off as recovery is limited to diffusion. Potential to be used in conjunction with containment treatments
	De-acidise soil	**Likely**	Established cheap technology
	Thermal treatment	**Intermediate**	Costs high and not for CO_2 plume but clean-up of the trace elements potentially mobilised by the CO_2 contamination
	Capping	**Likely**	Cost will depend on extent of cap and most likely to be used in conjunction with a treatment.
	Gas collection trench	**Likely**	Cheap and established method to collect soil gas.
	Ecosystem restoration	**Likely**	Final result of any contamination clean up.
Bioremediation	Bioremediation of low pH groundwaters	**Intermediate**	Cheap established option, but extent controlled by ideal biological condition.
	Bioremediation of CO_2	**Minor**	Cheap, extent controlled by ideal biological condition. But CO_2 specific microbes sill to be field tested.
	Bioremediation of toxic metals	**Intermediate**	Cheap established option, but extent controlled by ideal biological condition.
	Bioremediation of hydrocarbons	**Intermediate**	Cheap established option, but extent controlled by ideal biological condition.
	Natural attenuation	**Likely / Intermediate**	May be first step in the risk assessment procedure, however high costs associated with monitoring.
Buildings	Passive vapour intrusion mitigation	**Likely**	Established cheap technology
	Passive / active sub slab venting	**Likely**	Established cheap technology
	Active vapour intrusion mitigation – subsurface pressurisation	**Likely**	Established cheap technology
	Block wall depressurisation	**Likely**	Established cheap technology
	Active ventilation	**Likely**	Established cheap technology
	Passive ventilation	**Likely**	Established cheap technology
	Demolish and rebuild to	**Minor**	Final resort if other building

	suitable standards.		remediation technologies are unsatisfactory.

Table 3 indicates that there are a wide range of remediation techniques available for near surface CO_2 remediation and that any remediation strategy will be site specific.

5. Conclusions

For a hypothetical leak from a CO_2 storage site, a wide variety of potential remediation techniques are available, 'off the shelf', that could be applied. Many such techniques have been developed by the pollution clean-up industry, but very few have been developed, or even tested, for the specific case of a CO_2 leak. However, commercial companies exist that have extensive expertise in pollution clean-up, and contracting work to one of these companies would seem to be a sensible step in the event of an on-shore leak. The techniques that might be employed are very site- and scenario-specific, making generalizations difficult. An essential first step in any clean-up procedure is a comprehensive site study, and desk-top evaluation of all the available techniques to select the most appropriate. The aim of the remediation must also be clearly identified. The likely cost of remediation is very difficult to estimate, partly because very few figures for cost are available from the literature, and partly because the costs will be very dependent upon the leakage scenario and the purpose of the remediation.

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